Cesium Management and Disposition for the Low Activity Waste Pretreatment System (LAWPS)

Summary

The Hanford Advisory Board, following lengthy discussions and reviews conducted by the Board’s Tank Waste Committee with the U.S. Department of Energy (DOE) Office of River Protection (ORP), has completed a review of the proposed Direct Feed Low Activity Waste (DFLAW) process and the Low Activity Waste Pretreatment System (LAWPS). Specifically, the Committee’s discussions centered on the proposed management and potential disposal paths of the High Level Cesium Waste resulting from the LAWPS process. This review was performed at the request of DOE-ORP Federal Project Director, Low Activity Waste Pretreatment System, as described in the Hanford Advisory Board 2015 and 2016 Work Plans. Specific areas to be discussed in this work plan item included:
- Are there alternate cesium removal, storage, and disposition technologies that should be considered under Direct Feed Low Activity Waste scenarios?
- What would be the implications for long term cleanup planning on the Central Plateau?

Background

Current DFLAW plans call for using an ion exchange process in the LAWPS to strip high level waste constituents, primarily highly radioactive cesium, from a waste stream from the tank farms creating a low activity waste feed for vitrification in the Low Activity Waste (LAW) facility. The cesium or high level waste would be returned to the existing waste tanks in the tank farms for later processing when the capability to process High Level Waste (HLW) in the Waste Treatment and Immobilization Plant (WTP) is available.

On September 24, 2013, the U.S. Department of Energy (DOE) released the Hanford Tank Waste Retrieval, Treatment, and Disposition Framework (Framework) document. This document describes a strategic framework for addressing the risks and challenges to completing the DOE Office of River Protection (ORP) mission by implementing a phased approach that would:
- Begin immobilization of the tank waste as soon as practicable through the Direct Feed Low Activity Waste (DFLAW) process.
- Process transuranic (TRU) tank wastes for disposal at the Waste Isolation Pilot Plant (WIPP).
- Resolve technical issues for the Pretreatment (PT) and High-Level Waste (HLW) Facilities, including determining how to adequately mix and sample the waste prior to processing, to enable design completion, and the safe completion of construction, startup and operations of these facilities.

Immobilization of the approximately 56 million gallons of radioactive and chemical wastes stored in 177 underground tanks located on Hanford’s Central Plateau will occur in the Waste Treatment and
Immobilization Plant (WTP). The complexity of both the waste itself as well as the WTP facilities has led to difficult, and to date, unresolved technical issues for the portions of the facility (PT Facility and to a much lesser extent the HLW Facility) that will process the solid portions of the waste. Because the current design of WTP anticipates that all waste will be processed through the PT Facility, immobilization of any waste could not occur per the current plan until the many technical issues involving the PT Facility are resolved. Therefore, an alternative approach for immobilizing waste as soon as practicable, while simultaneously resolving the remaining technical challenges, was identified. By adopting a DFLAW option in which the waste bypasses the PT Facility, waste immobilization could begin significantly earlier than if treatment of the waste is delayed until all technical issues are resolved and the PT and HLW Facilities are completed.

The Framework document divided the 56 million gallons of tank waste into three major categories for treatment:

1. Low-activity waste;
2. Potential contact-handled transuranic waste (CH-TRU); and
3. High-level waste, which is further subdivided into waste not requiring special handling (easier to process) and waste requiring special handling (harder to process).

The low-activity waste consists primarily of the supernate (liquid) portion of the tank waste with most of the solids and radioactivity removed before vitrification, low-activity waste will be the largest tank waste stream by volume (approximately 90% of the volume), but the lowest in radioactivity content (approximately 10% of the curies). Since the low-activity waste makes up approximately 90% of the total volume of waste to be treated, and has the greatest influence on the total duration of the Hanford tank waste mission. The liquid form of this waste makes it susceptible to leakage. The low activity waste is also the tank waste most easily processed through the WTP. In particular, at the present time it is felt that there are no significant technical risks associated with vitrifying this waste stream in the LAW Facility.

Beginning LAW Facility operations before the PT Facility is operational would require a capability to remove the cesium and small amounts of transuranic and strontium-90 solids from the liquid supernatant waste stream so that low-activity waste could be directly fed to the LAW Facility for glass immobilization.

ORP’s analyses of this approach indicates that a standalone Interim Pretreatment System Facility would best address this need. It would be located between the tank farms and the LAW Facility and would remove the solids and cesium from the liquid waste stream. In addition some space has been set aside to possibly remove other radioactive elements or test improvements in currently planned separation techniques. This facility would provide the processing capability to support a DFLAW operation prior to the completion of PT. As this option uses mature technologies, it is felt that the technical risks associated with this alternative are low.
The return of radioactive cesium to the tanks has several disadvantages such as:

Cesium gamma emissions are the principle radiation hazard to the work force.

- Cesium makes up about 50% of hydrogen generated in tanks/Pretreatment. 7 of 12 tanks scheduled for DFLAW have high hydrogen generation rates.
- Cesium places more radiation/heat stress on tanks; some nearing their design life.
- Cesium return to the DSTs is more expensive and creates more waste. It is cheaper in the short run and more costly in the long run.
- Returning cesium takes up tank space; less free DST space created.

The DFLAW will require the supernate stream to be transferred to the LAW Facility for vitrification following interim pretreatment. Secondary liquid wastes generated from the LAW Facility off gas system and LAWPS facility effluent would be transferred to a new Effluent Management Facility for volume reduction evaporation, and sent back to the double shell tank farms. Alternatively LAW Faculty off gas and LAWPS effluent could be transferred to the tank farms and volume-reduced through evaporation activities using the existing 242-A Evaporator. Waste streams from LAWPS would be primarily from resorcinol formaldehyde cesium eluent (which would be neutralized and likely sent directly back to DST), and water solutions of: 0.4% sodium hydroxide, 4% sodium hydroxide, 2.8% nitric acid, deionized water, and rinses of spent resin before disposal. These total transactions for every 1 million gallons of DST supernatant removed from the tanks and processed approximately 0.33 million gallons are returned to DSTs after evaporation. So for every 3 parts of supernatant removed and process 1 part of waste is returned to the DSTs.

DST space analysis of DFLAW in a little more depth:

The actual space taken up by returning neutralized cesium eluded off the LAWPS cesium resin back into the DSTs is ~9% of the supernatant volume removed. This does not include concentration by evaporation which is probably not the choice operation for many reasons. The approximate remaining 24% volume returned to the DSTs (remember 3 parts volume removed from DST, 1 part volume retuned to DSTs) is from LAWPS resin pretreatment, LAWPS resin post cesium elution reactivation, and from LAW off gas processes. This 24% of volume returned to the tanks includes significant reduction by evaporation by a factor of ~2.5 or slightly more. The large majority of this is form LAW off gas. (Numbers to be provided).

As requested by ORP in the Hanford Advisory Board (Board) 2015 and 2016 Work Plan, the Board has conducted an in-depth review of the preliminary design associated with the DFLAW and the possible alternate cesium removal, storage, and disposition technologies that might be considered for use in the DFLAW. The following discussion summaries the options that the Board considered and our findings and recommendations.
Cesium Disposition Alternatives considered

The alternatives were developed and considered for the disposition of the Cesium removed from the waste steam as part of the DFLAW process.

Option:

1. Return Cesium back to the DST (current DOE baseline alternative)

2. Process DFLAW baseline with cesium returned back to DSTs and expedite Direct Feed HLW. For DFLAW initial runs process low concentration cesium supernatant only (no saltcake in initial runs). Initial process sequence for DFLAW tanks is: 1st tank AP-104, 2nd tank AP-106, 3rd tank AP-108, and 4th tank AP-108. Expedite Direct Feed HLW process by installing a 100,000 gallon below ground DST tank with some solids/liquid separation capability, large single (replaceable) mixing impeller, hard installed sampling ports, small sampling and ventilation support building, and related underground piping. All tank sludge solids and some related saltcake retain in sludge transfers are to be process by Direct Feed HLW without any pretreatment extraction of any kind for the life of the mission. Once Direct Feed HLW is operational direct all cesium from LAWPS process to HLW glass. Once Pretreatment Facility is complete integrate off gas of all glass plants and processes and expose Pretreatment Facility to minimal entrained solids of if absolutely necessary for some select HLW glass batches use PT to process sludge solids.

3. Grout Cesium and dispose in a Licensed Commercial Disposal Facility using LAWPS current resorcinol formaldehyde baseline resin process. The Federal LLRW site at Texas WCS has a current maximum Curie limit of 5.6 MCi. WCS can currently accept ~2.8 MCi of cesium-137 due to barium-137m progeny. This equates to 608 cubic meters of Class C waste at maximum cesium-137 concentration LLW Class C.

4. Grout Cesium with non elutable media and dispose in a Licensed Commercial Disposal Facility. Use a non-elutable media contained within the inner layer of the final waste disposal package. Media/extraction package will be porous enough to be grouted with extra fine grout in place after extraction and water flush. Media will be tailored to only bind cesium to give LLW C waste. (There is are several composite matrixes that can do this and are used or have been used especially in Eastern Europe and LLNL. They have better cesium selectivity than resorcinol formaldehyde resin, are high pH compatible, and there is no resin regeneration waste sent to tanks. The Federal LLRW site at Texas WCS has a current maximum Curie limit of 5.6 MCi. WCS can currently accept ~2.8 MCi of cesium-137 due to barium-137m progeny. This equates to 608 cubic meters of Class C waste at maximum cesium-137 concentration LLW Class C.

I will expound on option 2, 3, and 4 more. Option 2 is the best from an economic, regulatory, and stakeholder perspective. It is support by ORP management and the Nez Perce Tribe, but does not have a full economic analysis. DOE will probably take 2 years to do the math. May first
pass was $5-8B in savings. After talking with the glass scientists and seeing more of the whole plan the saving is ~$10B.

Some combination of 2 and 3 or 2 and 4 is also possible. But this sucks money from DF HLW which is the better win.

Option 4 is viable only in context. It is my understanding that DOE has no current supplier of the spherical resorcinol formaldehyde resin of the type used in all their tests. That may be a good thing because the resin manufacturing method can be made better with significantly less swelling on going to sodium form by slightly modifying the current manufacturing recipe. So option is not that hard to accept either.
Option 1 - Return Cesium back to the DST (current DOE baseline alternative)

The solids and cesium and possibly other radioactive elements will be removed from the liquid waste stream from the tank waste prior to vitrification in the LAW Facility. The Cesium is captured using Ion Exchange Resin media, then eluated with nitric acid, neutralized and returned to the DST. Secondary liquid wastes generated from the LAW Facility offgas system would then be treated and volume-reduced through evaporation activities using the existing 242-A Evaporator in the tank farms.
Option 2 - Dispose of Cesium in deep geologic Bore Holes

The solids and cesium and possibly other radioactive elements will be removed from the liquid waste stream from the tank waste prior to vitrification in the LAW Facility. The Cesium is captured using Ion Exchange Resin media, then eluated with nitic acid, neutralized, and treated and packaged for disposition in a deep geologic Bore Hole. Secondary liquid wastes generated from the LAW Facility offgas system would then be treated and volume-reduced through evaporation activities using the existing 242-A Evaporator in the tank farms.
Option 3 - Grout Cesium and dispose of at Licensed Commercial Disposal Facility

The solids and cesium and possibly other radioactive elements will be removed from the liquid waste stream from the tank waste prior to vitrification in the LAW Facility. The Cesium is captured using Ion Exchange Resin media, then eluated with nitric acid, neutralized, and grouted and packaged for disposition in a Licensed Commercial Waste Disposal facility. Secondary liquid wastes generated from the LAW Facility offgas system would then be treated and volume-reduced through evaporation activities using the existing 242-A Evaporator in the tank farms.
Option 4 - Store Cesium in Ion Specific Media for Future Federal Disposal

The solids and cesium and possibly other radioactive elements will be removed from the liquid waste stream from the tank waste prior to vitrification in the LAW Facility. The Cesium is captured using a non-elutable Ion Specific media, the module containing the Cesium is then vitrified in a modular vitrification melter and stored for future disposal in federal repository. Secondary liquid wastes generated from the LAW Facility offgas system would then be treated and volume-reduced through evaporation activities using the existing 242-A Evaporator in the tank farms.
Reasons for Not Returning Cesium to Tanks from DFLAW

- Cesium is present in HLW mostly in salt cake and supernatant as stable Cs-133 and radioactive Cs-134, Cs-135, and Cs-137. Cs-134 has mostly decayed away, leaving Cs-135 and Cs-137. Cs-137 decays to Ba-137m which decays to Ba-137. This is the principle gamma source in tanks. There is ~3 to 4 times more total cesium verses Cs-137 radioactivity.

- Reasons for not return cesium to tanks, because Cs-137 is:
  - Principle RAD hazard for work force.
  - Makes up about 50% of hydrogen generated in tanks/Pretreatment. 7 of 12 tanks scheduled for DFLAW have high H₂ generation.
  - Places more RAD/heat stress on tanks; some nearing their design life.
  - Cost more money and creates more waste. Cheaper in the short run/more costly in the long run.
  - Returning cesium takes up tank space; less free DST space created/ return 1 Mgal for every 3 Mgal removed from DSTs.

Several Possibilities for Cesium Pathway Using Non-Elutable Resin, Package, and Disposal; RPP-RPT-57115

- Alternative 1: Use non-elutable CST resin and ship to deep geological repository as HLW. Not likely a solution since repository is not operational. This could change hopefully by time HLW glass is being made.

- Alternative 2: Use low loading zeolite exchange media, ship to WCS Texas, requires WIR. Class C waste because of Cs-137. Nevada National Security Site is the other option but more DOE red tape (paperwork) is required.

- Alternative 3: IDF disposal as in Alternative 2, not in TC&WM EIS/ROD so amendment required and WIR required. 2-4 of 12 DFLAW tanks spent resin is likely TRU, but don’t tell Texas.

Cost for 5 years operations with resin disposal, $Million

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Cesium Pathway in Processes Using Non-Elutable Resin with Glass Stabilization and Elutable Resin with Less Waste

- Another alternative: Kurion and SRNL are proposing a modified Kurion system for cesium removal and interim storage in support of direct feed LAW. Details to be discussed. Likely a crystalline silicotitanate exchange media that is easily incorporated into glass, Geo Melter.

- Even another alternative: proposal to use Electro-active ion exchange by the Nez Perce Tribe. Used conductive ion exchange media of similar structure and selectivity to current resin. Does not require nitric acid and sodium hydroxide for regeneration and you get a counter-ion for free, technetium-99. Resin unloading in accomplished by switching electrical voltage opposite loading voltage. Waste cones off very concentrated giving small storage volume or borehole-able package sizes (grout). This is not electrodeionization (EDI). Electro-active ion exchange has not been evaluated much for nuclear waste, 1 or 2 minor examples. Could not be a mainstream proposal without proof of concept on Hanford waste types.

**Next Steps, Considerations**

- Waste from ion exchange and spent elutable or spent non-elutable resin will be Class C, GTCC, CH-TRU, RH-TRU, and/or HLW.

- Need to determine approximate fraction of expected waste destinations and types for proposals.

- Need to determine basic short term and long term costs for the above disposition destinations. On a cost basis the results currently are: HLW deep geological repository = 2 X WCS = 10-72 X IDF. IDF looks unrealistically cheap.

- Need to determine cost and destination of deep boreholes. Is DOE serious about this and do they have a regulatory pathway which is achievable?

- Look at site impacts and see what pathways can minimize impacts. Rank depositions: HLW = RH-TRU = Deep Borehole > GTCC > Class C

- Out of the box thinking: Use current ORP plan and designate a DST for Cs-137 waste or build one with significant cooling that could handle it.